CALCULATION OF ADVECTIVE MASS TRANSPORT IN HETEROGENEOUS MEDIA

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ABSTRACT. A coupled analytical/numerical procedure for prediction of solute transport in heterogeneous media is described. The procedure consists of an analytic solution of the hydraulic equations, followed by a numerical solution for solute transport using the method of characteristics. The characteristics are determined by fourth-order Runge-Kutta and predictor-corrector algorithms. Accuracy of solute transport calculation is enhanced by the fact that fluid velocity can be directly obtained at *a priori* undetermined points in the flow field.

The solute transport process is considered to be entirely advective, neglecting the effects of mechanical dispersion and molecular diffusion. Evidence is presented to demonstrate that purely advective processes in both heterogeneous and homogeneous media can produce large "apparent dispersion." Such dispersion is shown to be easily capable of overwhelming any reasonable estimates of dispersion or diffusion based upon laboratory analyses of homogeneous media. For groundwater contamination problems, it is concluded that precise definition of the spatial variability of hydraulic properties is crucial to the accurate determination of the trajectory of contaminated waters.

BACKGROUND. At the scale of individual grains, the transport of a conservative solute through a porous medium is clearly an advective phenomenon. Solute particles are wafted along by fluid as it flows over tortuous routes in the general direction of the potential gradient. Close observation of the movement of initially adjacent solute particles would reveal their tendency to become separated. Contributing to the separation one would observe: (a) random bifurcation of pore channels, (b) a large range of fluid velocities across individual pores, and (c) differences of fluid velocity from one pore to another. To a very minor extent, pore scale advection is supplemented by molecular diffusion.

For a fluid of nonuniform concentration which saturates a porous medium, the separation of solute particles amounts to mixing, resulting in changes of local solute concentration. Buyevich et al. (1969) noted that the pore scale advective mixing process is very much like the mixing resulting from ordinary fluid turbulence.

The usual material continuum approach to porous media modeling defines solute transport in terms of macroscale mass fluxes (Bachmat and Bear, 1964). Although the entire process is fundamentally advective at the pore scale, the macroscale description of advection can represent only an average transport, strictly in the direction of the potential gradient. The pore scale mechanisms listed above as (a) through (c) cannot be accounted for by macroscale advection alone.

Dispersion is an additional macroscale solute flux whose purpose is to account for the pore scale mechanisms which cause mixing. Dispersive flux is assumed to be proportional to concentration gradient.

Harleman et al. (1963), Klotz and Moser (1974), and others have conducted laboratory experiments on columns of homogeneous media to determine the relative magnitudes of macroscale advection and dispersion. Their results are presented as a correlation between the magnitude of dispersion, the potential gradient, and the physical properties of fluid and media.

Many investigators (e.g. Pinder, 1973; Konikow and Bredehoeft, 1974) have applied macroscale advection-dispersion models to field problems. Calibration of these models generally leads to the assumption of dispersive fluxes which are orders of magnitude greater than would be expected on the basis of lab analyses of porous material samples. Gelhar et al. (1979) reiterate the conclusion that this discrepancy is related to local heterogeneity of porous medium hydraulic properties. The experimental results of Skibitski and Robinson (1963) substantiate this by illustrating the dominant effect of heterogeneity on the transport of dye in sand flumes.

THEORY. The aim of this paper is to demonstrate a coupled analytical/numerical technique for predicting the transport of conservative solutes in heterogeneous media. The approach presumes that genuine dispersion is negligible compared to true macroscale advection when that advection fully accounts for heterogeneity and nonuniform flow.

In order to accurately determine the effect of heterogeneity on macroscale advection, an analytic solution for hydraulic potential is obtained. Application of Darcy's law yields an analytic expression for average linear velocity which can be evaluated at *a priori* unspecified points $\vec{x}$. As part of the technique, medium properties are accounted for as known (or interpolated) explicit functions of $\vec{x}$. Given an accurate description of flow field, streamlines are calculated by applying the method of characteristics. Advection is determined from the rates of flow along the streamlines.

Example problems are used to demonstrate the fact that genuine dispersion can be easily overwhelmed by the effects of heterogeneity and nonuniform flow. In each example, flow is assumed steady and horizontal. The analyses apply to confined aquifers and also to phreatic aquifers where the Dupuit assumptions and linear approximation are valid.

Advection-dispersion equation (numerical solution). The control volume approach can be used to derive the advection-dispersion equation (Daly, 1979):

$$\phi \frac{\partial C}{\partial t} - \nabla \cdot (D \nabla C) + \vec{v} \cdot \nabla C = \frac{\tilde{C}}{\rho} + q \frac{(\hat{C} - C)}{\rho} \qquad (1)$$

where:

ϕ = effective porosity, dimensionless;
C = mass fraction of the pore fluid; the ratio of the mass of solute in a given volume to the total mass of fluid in that volume, dimensionless;
D = dispersion coefficient tensor, L^2/T;

$\vec{v}$ = specific discharge, L/T;

$\tilde{C}$ = contaminant mass source/sink strength representing the exchange of mass between fluid and porous matrix, M/L^3T;

ρ = fluid density, M/L^3;

q = recharged fluid mass strength, $M/L^3T \geq 0$;

$\hat{C}$ = mass fraction of recharged fluid, dimensionless.

Considering (1), it is clear that the effect of fluid withdrawals is not felt directly through the terms on the RHS of the equation. However, withdrawals do affect the transport by modifying the flow field (represented in (1) by $\vec{v}$).

If dispersion is neglected compared to macroscale advection, (1) can be written, for the simple case of no source/sink terms, as:

$$\phi \frac{\partial C}{\partial t} + v_x \frac{\partial C}{\partial x} + v_y \frac{\partial C}{\partial y} = 0 \qquad (2)$$

Application of the method of characteristics transforms (2) into the equivalent system of ordinary differential equations:

$$\frac{dx}{dt} = \frac{v_x}{\phi} \equiv f(x,y,t) \qquad (3)$$

$$\frac{dy}{dt} = \frac{v_y}{\phi} \equiv g(x,y,t) \qquad (4)$$

$$\frac{dC}{dt} = 0 \qquad (5)$$

Equations (3) and (4) are used to determine the trajectories (also called the characteristic lines) of fluid particles in the flow field. Equation (5) is simply a statement of the fact that in the absence of sources or sinks the concentration of fluid particles remains constant. It is important to note that (3), (4), and (5) are not independent. Equation (5) is only valid along the trajectories defined by the joint solution of (3) and (4).

Determination of the trajectories of fluid particles is done by numerically solving the linked system of (3) and (4). A fourth order Runge-Kutta technique is used to start the procedure which can be continued

by a more efficient predictor-corrector scheme. The numerical solution method starts at a point (x_o,y_o) at time zero. The concentration is defined at all such points by an initial condition. Using a time step Δt, successive points (x_n, y_n) along the trajectory of the particle which began at point (x_o, y_o) are obtained. The Runge-Kutta algorithm for accomplishing this is:

$$x_{n+1} = x_n + \frac{1}{6}(a_1 + 2a_2 + 2a_3 + a_4) \tag{6}$$

$$y_{n+1} = y_n + \frac{1}{6}(b_1 + 2b_2 + 2b_3 + b_4) \tag{7}$$

where:

$$a_1 = \Delta t\ f(x_n,y_n,t_n) \tag{8}$$

$$b_1 = \Delta t\ g(x_n,y_n,t_n) \tag{9}$$

$$a_2 = \Delta t\ f(x_n + a_1/2,\ y_n + b_1/2,\ t_n + \Delta t/2) \tag{10}$$

$$b_2 = \Delta t\ g(x_n + a_1/2,\ y_n + b_1/2,\ t_n + \Delta t/2) \tag{11}$$

$$a_3 = \Delta t\ f(x_n + a_2/2,\ y_n + b_2/2,\ t_n + \Delta t/2) \tag{12}$$

$$b_3 = \Delta t\ g(x_n + a_2/2,\ y_n + b_2/2,\ t_n + \Delta t/2) \tag{13}$$

$$a_4 = \Delta t\ f(x_n + a_3,\ y_n + b_3,\ t_n + \Delta t) \tag{14}$$

$$b_4 = \Delta t\ g(x_n + a_3,\ y_n + b_3,\ t_n + \Delta t), \tag{15}$$

and f and g are defined in Equations (3) and (4).

Runge-Kutta algorithms belong to the set of self-starting numerical solution methods. Self-starting means that the determination of all successive points (x_n, y_n) requires only the starting point (x_o, y_o). In other words, calculation of x_n and y_n depends only on the known values x_{n-1} and y_{n-1}. The set of non self-starting methods require the values (x_n, y_n) to be given at more than one point along the trajectory. For example, a fourth order predictor-corrector algorithm called Milne's method requires the values x_o, x_1, x_2, x_3, y_o, y_1, y_2, y_3 to calculate successive values of x_n and y_n.

Beside the question of starting values, the efficiency of the calculation procedure is an important factor in selecting a numerical method. It turns out that Milne's algorithm is significantly more efficient than the Runge-Kutta method, although both are fourth-order accurate. One numerical procedure proposed in this paper is that which takes advantage of the Runge-Kutta self-starting feature and the efficiency of Milne's method. Given a starting point (x_o, y_o), the Runge-Kutta procedure is used to obtain (x_1, y_1), (x_2, y_2), (x_3, y_3). At that stage the necessary starting values are available for Milne's method which is then used to generate succeeding points.

Non self-starting methods typically assume constant Δt, whereas self-starting methods allow for change of Δt at each time step. For problems in which the frequent change of Δt is desirable, exclusive use of a self-starting method, such as the Runge-Kutta algorithm, is advised.

Milne's predictor-corrector method consists of two steps. First, predicted estimates of x_{n+1} and y_{n+1} are calculated. Let these be denoted x^*_{n+1} and y^*_{n+1}. Second, the predicted values are corrected to obtain the final values x_{n+1} and y_{n+1} at the end of a time step. The algorithm is :

$$x^*_{n+1} = x_{n-3} + \frac{4\Delta t}{3} \left[2x'_n - x'_{n-1} + 2x'_{n-2}\right] \tag{16}$$

$$y^*_{n+1} = y_{n-3} + \frac{4\Delta t}{3} \left[2y'_n - y'_{n-1} + 2y'_{n-2}\right], \tag{17}$$

then:

$$x_{n+1} = x_{n-1} + \frac{\Delta t}{3} \left[x^{*\prime}_{n+1} + 4x'_n + x'_{n-1}\right] \tag{18}$$

$$y_{n+1} = y_{n-1} + \frac{\Delta t}{3} \left[y^{*\prime}_{n+1} + 4y'_n + y'_{n-1}\right] \tag{19}$$

where:

$$x'_n = f(x_n, y_n, t_n) \tag{20}$$

$$y'_n = g(x_n, y_n, t_n) \tag{21}$$

Consideration of the Runge-Kutta and the Milne algorithms shows that Milne's method requires only two evaluations of f and g per time step, whereas Runge-Kutta requires four. This makes Milne's method more efficient.

Steady flow between a source/sink pair. Consider the steady flow of fluid between a line source and a line sink of equal strength = Q, separated by a distance = a. Let the source/sink pair be located in a homogeneous, isotropic medium of infinite extent, and saturated thickness

b. Suppose that at time zero the concentration of solute at the source is changed from zero to C_o.

The well-known time dependent solution for the concentration of fluid recovered at the sink depends on the travel time of fluid particles (e.g. Charbeneau and Street, 1979). Travel time t is expressed as a function of θ, the direction of travel of a particle as it issues from the source. If the angle θ is measured from a line between the source and sink, then:

$$t = \frac{\pi b \phi a^2}{Q} \frac{1}{\sin^2\theta} [1 - \theta \cot\theta] \qquad \theta > 0 \tag{22}$$

where ϕ is the effective porosity. For $\theta = 0$, the minimal travel time t_m is:

$$t_m = \frac{\pi b \phi a^2}{3Q}. \tag{23}$$

At time $t > t_m$ the relative concentration of recovered fluid is:

$$\frac{C}{C_o} = \frac{1}{\pi} \theta(t) \tag{24}$$

where the function $\theta(t)$ is defined by (22).

The analytical solution to the source/sink problem was compared with a numerical solution obtained via the method of characteristics and the Runge-Kutta algorithm. Variables were assigned the values: a = 500 m, Q = 10000 m^3/days, b = 50 m, ϕ = 0.2. The numerical calculation began with Δt = 0.05 day; subsequent values of Δt were selected so as to allow fluid particles to travel about 10 meters per time step. Both analytical and numerical results are plotted in Figure 1; note that the two solutions practically coincide.

Since the concentration of fluid recovered at the sink varies with time, the transport may be viewed as a mixing process. In fact, this mixing or "apparent dispersion" is obviously just the result of nonuniform flow. The source/sink example leads to the conclusion that unrepresented nonuniform flow may result in considerable unexplained "dispersion."

Transport in heterogeneous media. Consider the transport of a conservative solute in a two dimensional steady flow field. The flow domain is assumed to be rectangular, L by H, having the distributions of transmissivity and effective porosity as shown in Figures 2 and 3.

Solution of the transport problem begins with the determination of flow field, which in turn begins with finding the hydraulic potential. Using the linearized Boussinesq equation, Daly and Morel-Seytoux (1981) determined an analytic solution to this problem subject to the boundary conditions on the potential h:

$h(0,y) = 0 \qquad h(L,y) = \lambda$

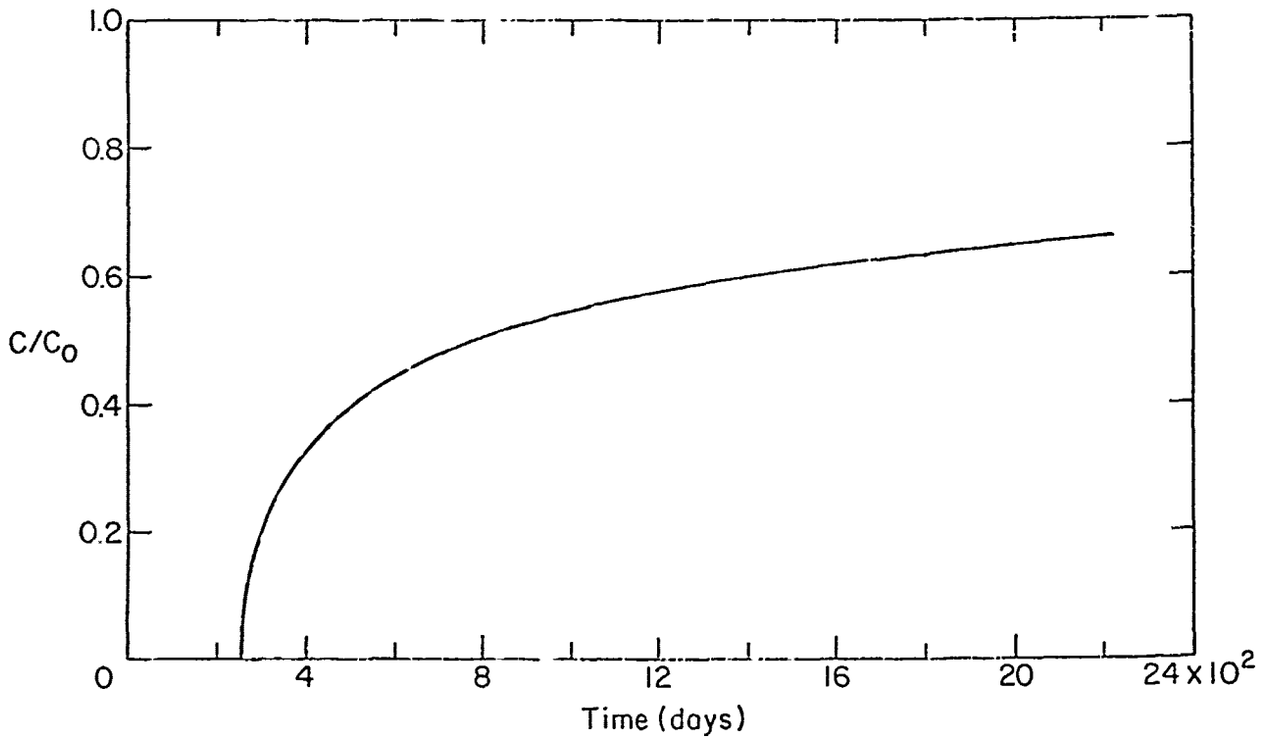

Figure 1. Relative concentration of fluid at the producing well of a source-sink pair.

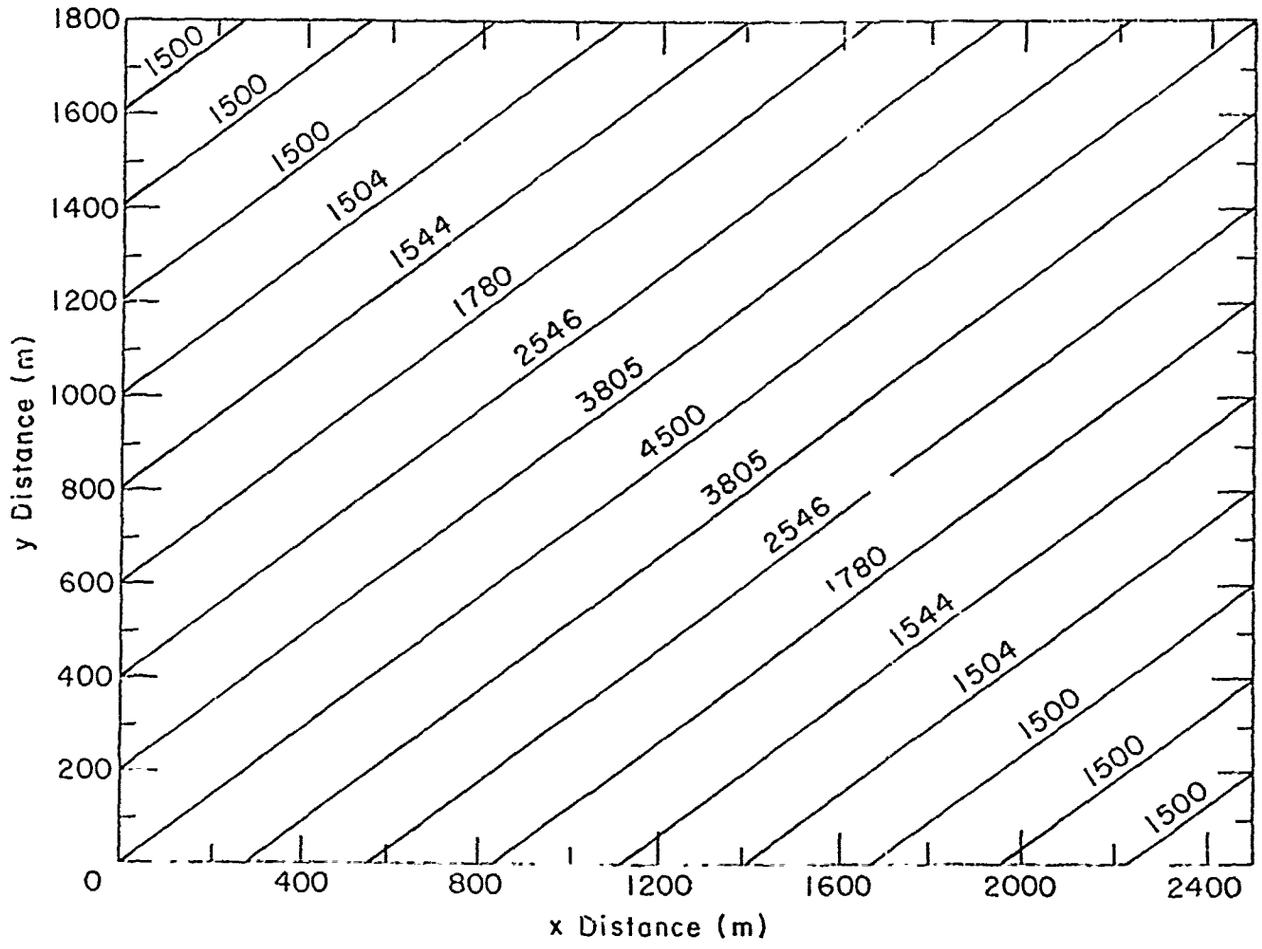

Figure 2. Distribution of aquifer transmissivity [m^2/day].

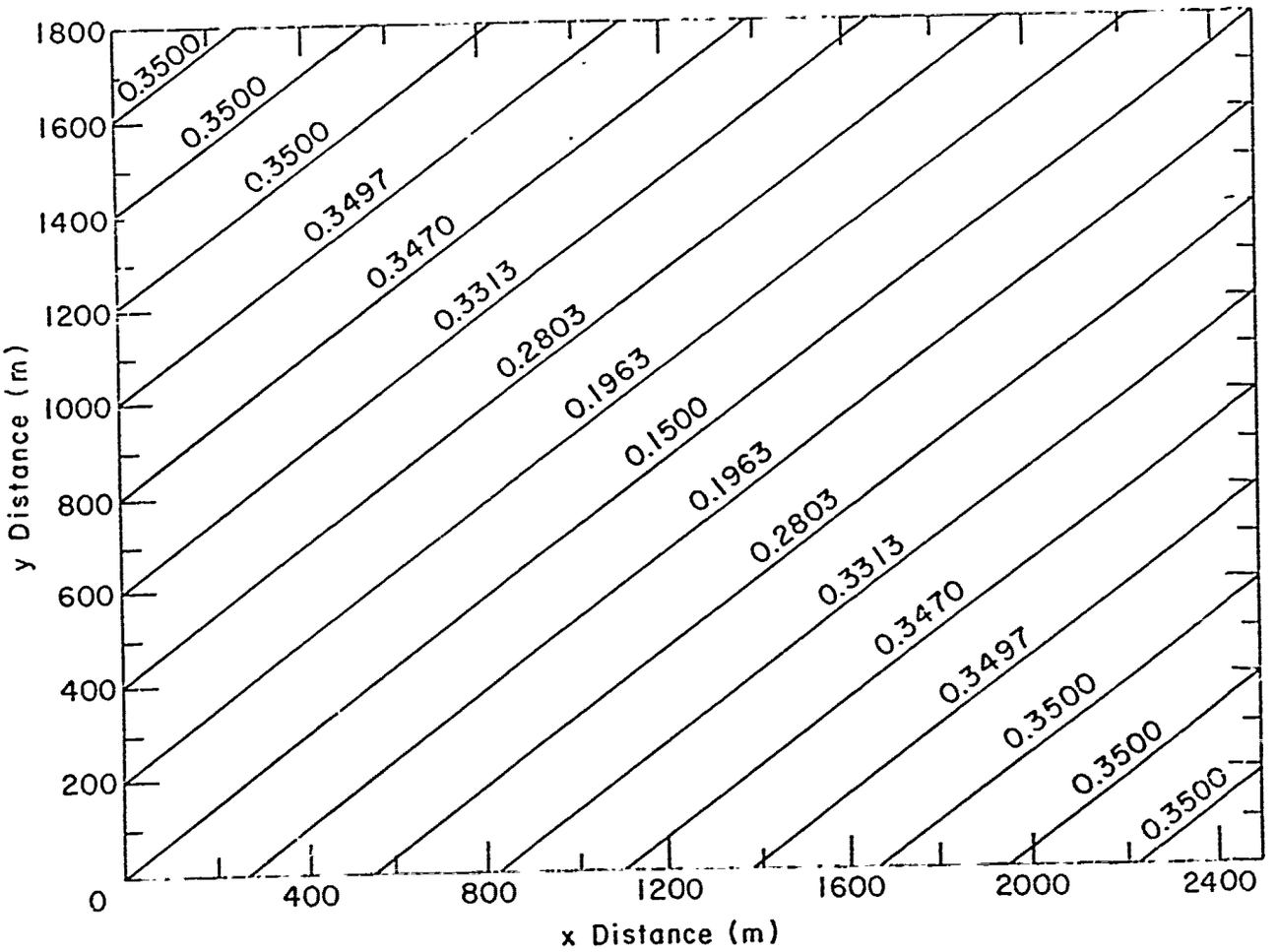

Figure 3. Distribution of aquifer porosity [dimensionless].

$$\frac{\partial h}{\partial y}(x,0) = \frac{\partial h}{\partial y}(x,H) = 0 \qquad (25)$$

Their solution is:

$$h(x,y) = \frac{2}{LH} \sum_{n=1}^{N} A(n,0) \sin \frac{n\pi x}{L} + \frac{x\lambda}{L}$$

$$+ \frac{4}{LH} \sum_{m=1}^{M} \sum_{n=1}^{N} A(n,m) \sin \frac{n\pi x}{L} \cos \frac{m\pi y}{H} \qquad (26)$$

where the Fourier coefficients A, dependent on medium heterogeneity, are found by the application of an integral transform method.

The specific discharge (and average linear velocity) associated with the potential distribution of (26) is obtained from Darcy's law. Equation (26) is easily differentiated to yield the hydraulic gradient. For the problem presented here: L = 2500 m, H = 1800 m, λ = 10 m, and saturated thickness is assumed constant and equal to 50 m. Figure 4 is a vector diagram of specific discharge.

The trajectories of fluid particles (located initially along the right hand vertical edge of Figure 4) were calculated by the Runge-Kutta, predictor-corrector method. A constant time interval of 100 days was used. The calculated trajectories define the streamlines shown in Figure 5; triangles are used to locate particles at 100-day intervals.

The movement of a sharp concentration front through the medium is shown in Figure 6. It is assumed that at time zero the concentration of fluid along the right hand boundary was instantaneously changed from zero to C_o. In the figure, the front is plotted at 200-day intervals. The movement of any particular point on the front is found by following that point along its associated streamline.

Consider the fluid which exits the porous medium at the left hand side of Figure 4. The average concentration of that fluid can be determined by calculating the time of breakthrough of many individual stream tubes. The ratio of the outflow produced by the tubes which have broken through to the total outflow can be obtained at any time. That ratio gives the relative concentration of the fluid flowing out of the medium. Using many more stream tubes than are shown in Figure 5, the calculation procedure was performed. The result is plotted as the solid line in Figure 7 (the "observed" breakthrough curve).

Using dispersion to account for the shape of the breakthrough curve. Suppose that the existing heterogeneity of the preceding problem is unknown. Suppose also that an experiment is conducted to determine a breakthrough curve; the result is the "observed" breakthough curve of Figure 7.

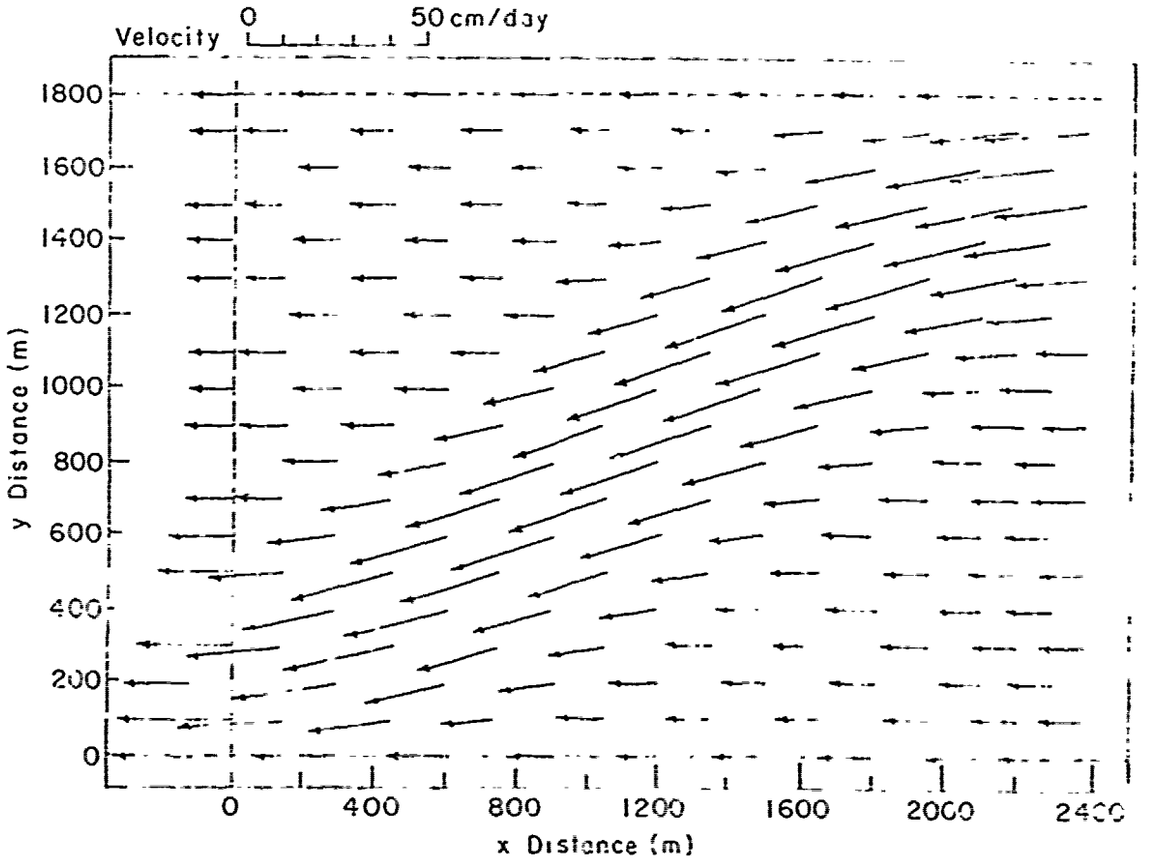

Figure 4. Specific discharge; flow pattern for two dimensional steady state flow.

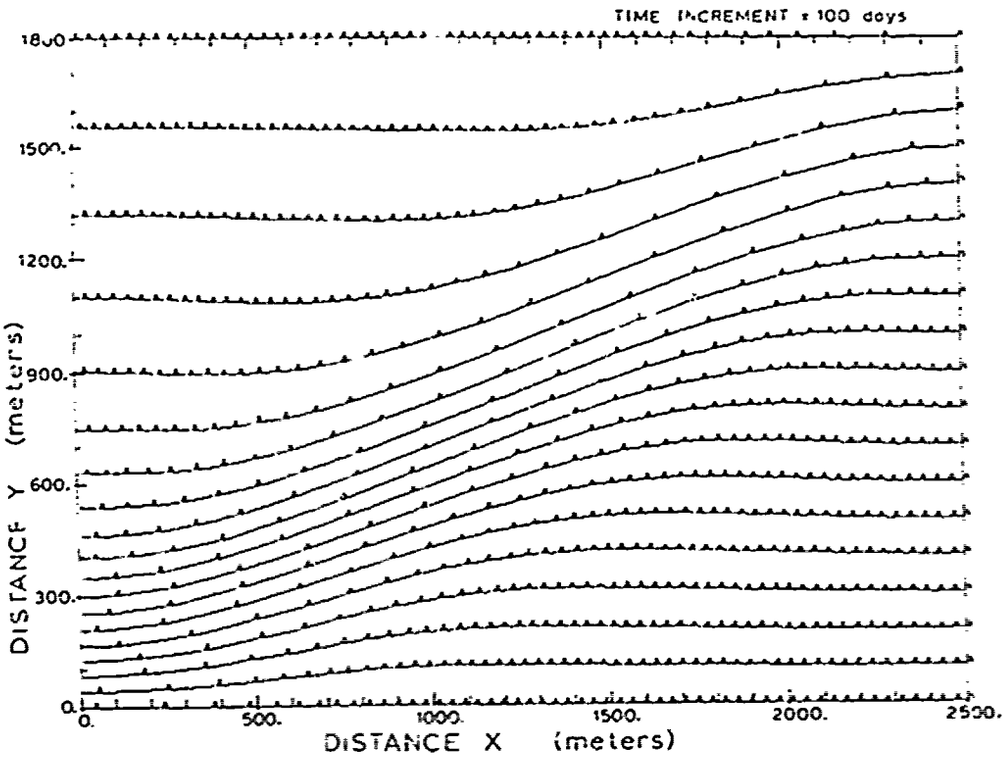

Figure 5. Streamline pattern for the flow field of Figure 4.

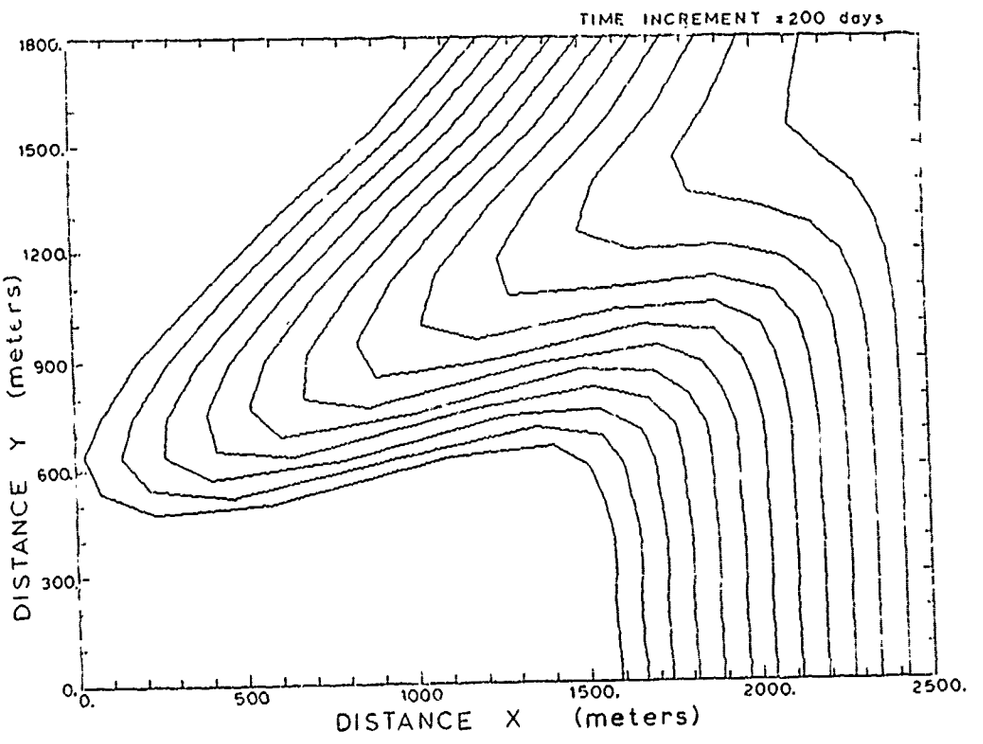

Figure 6. Movement of a solute front in the flow field of Figure 4.

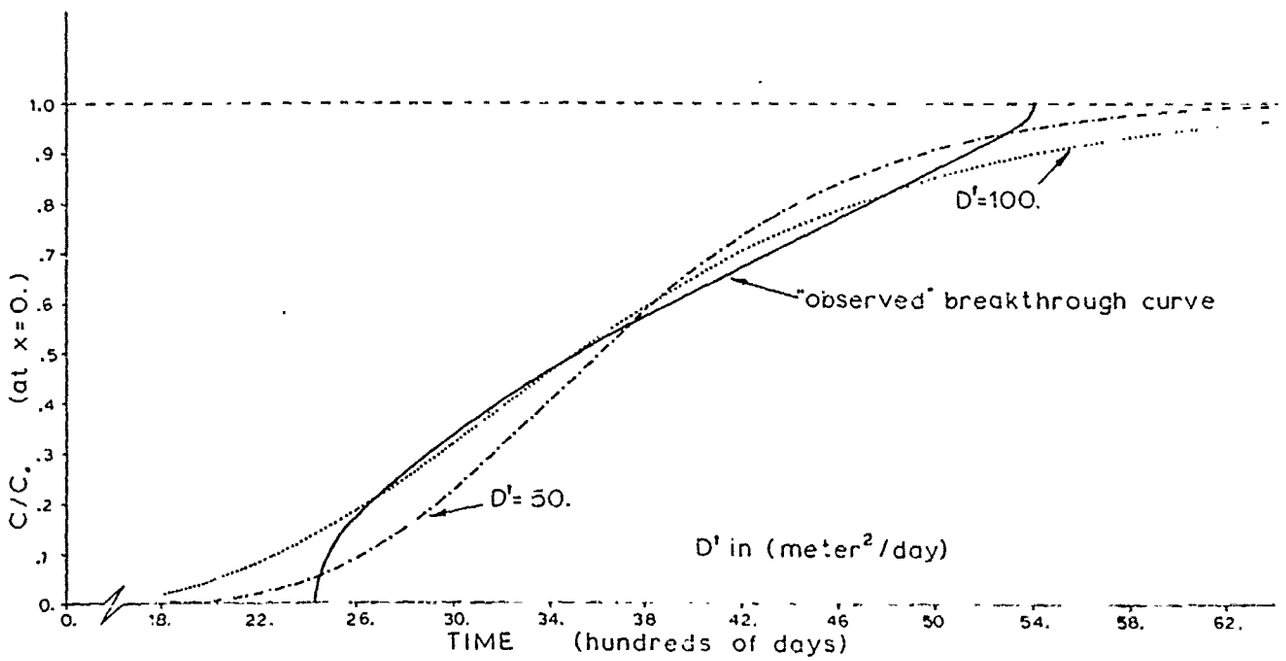

Figure 7. Breakthrough curves for the moving solute front of Figure 6.

If the porous medium were assumed homogeneous, all streamlines in Figure 5 would be parallel to the x axis. Flow would be steady and uniform and the problem could be considered one dimensional. A solution to the one dimensional advection-dispersion equation for steady flow has been obtained by Ogata and Banks (1961). The differential equation is:

$$\frac{\partial C}{\partial t} - D' \frac{\partial^2 C}{\partial x^2} + u \frac{\partial C}{\partial x} = 0 \qquad u \geq 0 \tag{27}$$

subject to:

$$C(0,t) = C_o; \quad C(\infty,t) = 0 \tag{28}$$

and the initial condition:

$$C(x,0) = 0 \tag{29}$$

where u = average linear velocity; $D' = D/\phi$. After a change of origin, the solution for the breakthrough curve is: at x = 0:

$$\frac{C}{C_o} = \frac{1}{2} \operatorname{erfc} \frac{L-ut}{2\sqrt{D't}} + e^{\frac{uL}{D'}} \operatorname{erfc} \frac{L+ut}{2\sqrt{D'T}} \tag{30}$$

For the problem considered here L = 2500 meters and

$$u = \frac{v}{\phi} = 0.6745 \text{ m/day} \tag{31}$$

Several estimates of the coefficient D' can be made. The resulting breakthrough curves are plotted in Figure 7 for the estimates D' = 50 and 100 m^2/day. Note that these two curves give an approximate fit to the observed breakthrough curve.

In a series of experiments dealing with one dimensional dispersion, Harleman et al. (1963) correlated dispersion coefficient with flow and media properties. A variety of unconsolidated materials were used. The flow and transport problem were such that the analysis of Ogata and Banks (1961) could be applied. Determining the breakthrough curve and the average linear velocity gave Harleman et al. the ability to estimate D' from Equation (30). Their correlation formula predicts for sand grains:

$$\frac{D'}{\nu} = 0.90 \left(R_{d_{50}}\right)^{1.2} \tag{32}$$

where: ν is the kinematic viscosity $[L^2/T]$,

$$R_{d_{50}} = \frac{|u| d_{50}}{\nu} \tag{33}$$

and d_{50} is the 50% grain size of the porous material.

If the shape of the observed breakthrough curve of Figure 7 is assumed to be the result of dispersion, (32) can be used to estimate d_{50} for the

porous material. With $D' = 100$ m^2/day, $d_{50} = 49.32$ m; for $D' = 50$ m^2/day, $d_{50} = 27.68$ m.

CONCLUSIONS. The above results dramatically show that: (a) even modest heterogeneity of porous media properties cannot be properly accounted for by dispersion, and (b) genuine dispersion is easily overwhelmed by the effects of heterogeneity.

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